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# Erosion and high temperature oxidation resistance of new coatings fabricated by a sol-gel route for a TBC application

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**Abstract.** This paper examines the erosion and cyclic oxidation performance of novel thermal barrier coatings produced via the sol-gel route. The ceramic top coat, with a thickness of 5-80  $\mu\text{m}$ , was deposited via a sol-gel route onto standard MCrAlY and PtAl bond coats. In both the erosion and the cyclic oxidation tests it was found that the bond coat had a profound affect on the results. The erosion of the sol-gel coatings were compared to standard EB PVD and PS TBCs and were found to be significantly higher. The effect of aging (100 h at 1100°C) on the erosion rates was also evaluated and was found to increase the erosion rates. The information obtained from the erosion and cyclic oxidation tests have highlighted the need to develop and optimise the parameters for producing thicker coatings

## Introduction

The thermomechanical performances of Yttria stabilised Zirconia (YSZ) thermal barrier coatings strongly depend on its microstructure. Currently, the ceramic topcoat is deposited by two main techniques: the Electron Beam Physical Vapour Deposition (EB-PVD) or the plasma spray process. Both result in highly oriented microstructures; indeed, columnar coatings are characteristic of the EB-PVD process and the superposition of YSZ lamellas are a characteristic of the thermally sprayed coatings. Recently, a soft chemical route, the sol-gel process, has been developed in order to produce a topcoat with a non-orientated microstructure. A major difficulty was the fabrication of thick coatings from a liquid phase precursor at temperatures below 1200°C. This paper presents the first results concerning the mechanical characterizations of the sol-gel coatings obtained on substrates with different roughnesses. First, the erosion performance of those coatings was studied and the results compared to the standard EB-PVD and plasma sprayed TBCs. Following these erosion studies, the cyclic oxidation behaviour of the sol-gel coatings were examined.

### Details of the Material Systems Studied and Fabrication of Sol-Gel Thermal Barrier Coatings.

The sol-gel top coats were prepared by the combination of the dip-coating technique with the alkoxyde route [1-2]. The microstructure of the coatings is quite different compared to the columnar or lamellar oriented ceramic top coat structures normally associated with TBCs. Indeed, by using the sol-gel process, the YSZ material and the pore network are randomly oriented in the coating as shown in Fig. 1.

For the characterisation studies and the erosion tests, two bond coat systems were used; a plasma sprayed NiCrAlY bond coat, with a surface roughness between 10-15 $\mu\text{m}$  Ra, and a nickel aluminide bond coat fabricated using the Snecma vapour aluminising process (APVS). The substrate for these studies was Hastelloy X. Hastelloy X is a Nickel-Chromium-Iron-Molybdenum alloy with a good combination of oxidation resistance, ease of fabrication and high temperature strength. (47%Ni ; 22%Cr ; 9%Mo ; 18% Fe).

For cyclic oxidation studies, the performance of the system is known to depend on the bond coat/substrate system, thus a (Ni,Pt)Al bond coat was used on a single crystal superalloy substrate (AM1). The surface roughness of the two vapour aluminised bond coats was within 1 $\mu\text{m}$  Ra.

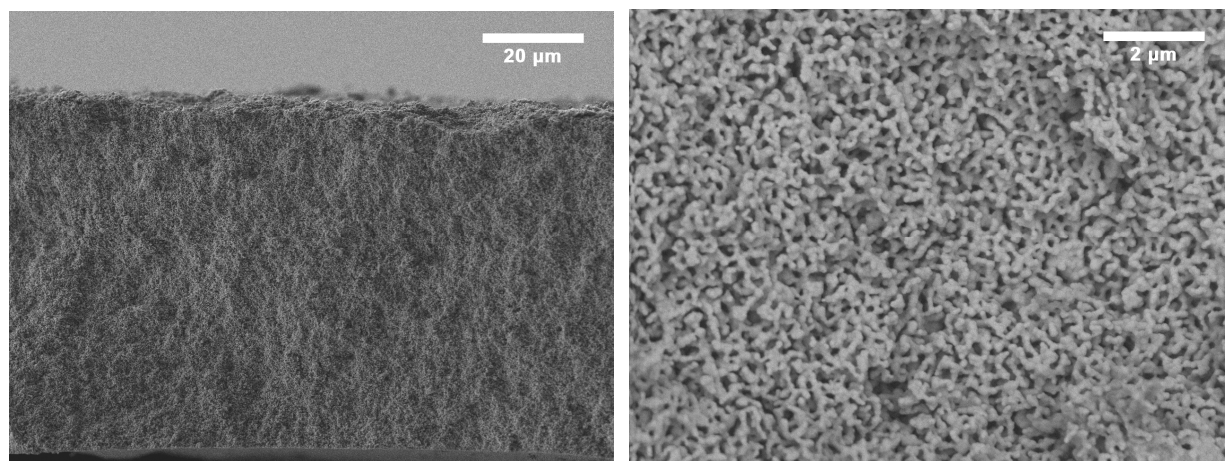


Fig. 1. Cross section micrographs of the random oriented YSZ top-coat obtained via the sol-gel route on the (Ni, Pt)Al/AM1 substrate, general view (left) and detail of the non oriented microstructure (right).

**Erosion testing.** All the erosion tests were carried out using the erosion facility at Cranfield University. A compressor supplies a pressure vessel and the air enters the acceleration tube. Then, the erodent material is fed via a screw feeder and a venture nozzle into this tube. The compressed air accelerates the particles before they impact the test specimen in the chamber. The test samples are attached to a carousel, which is capable of holding six different specimens at different angles with respect to the impacting particle stream.

All samples (reference TBCs, reference bond coats and sol-gel top coats) were weighed before the erosion testing and then placed in the carousel. Each sample was then exposed to the particle laden gas stream for 10s (short time exposure) or 30s (long time exposure), for reference samples, after which, and once all the samples had been tested, they were removed from the rig and re-weighed. This was repeated for six exposure periods or until coating penetration. Thus, the erosion rate can be reported as mass loss with the units g/kg, i.e. materials loss in grams per kilogram of erodent impacting the sample. The erodent, projected with a 1.5 bar pressure, was alumina particles with

diameter comprised between 63µm and 65µm. The tests were all conducted at room temperature and 30° impact for these experiments.

**Cyclic oxidation testing.** The cyclic oxidation tests were performed using a motorised cyclic oxidation furnace at CIRIMAT. Samples were placed on the motorised support and intermittently moved between a cool section, maintained at room temperature and a preheated vertical furnace at 1100°C. The hot dwell time was one hour, whereupon the samples were removed to the cool section and cooled to room temperature using a multi-channels air blast. All the tests were performed in laboratory air.

## Results and Discussions

**Characterization of the reference samples.** Table 1 presents the erosion rates obtained for the reference systems studied. Moreover the graph Fig. 2 shows the evolution of their mass loss versus the mass of erodent projected at their surfaces. At the beginning of the test, (up to about 0.5g of erodent), it can be seen that for each sample, the mass loss is not linear (Fig. 2), after which a linear mass loss is attained. This transient mode is associated with the preconditioning of the surface and of the subjacent impacted zone. Then, the evolution of the erosion rate becomes linear and it corresponds to the degradation of the coatings.

Comparing the behaviour of the reference TBCs, it can be seen that the plasma sprayed TBC system erodes at a faster rate than its EB PVD counter part. In this study the steady state erosion rate of the plasma sprayed TBC is twice that of the EB PVD when impacted at 30° impact angle. The transient erosion behaviour is much greater with the erosion rate for the rougher plasma spray TBC 8 times that of the EB PVD system. This increased transient behaviour is thought to be due to the difference in the initial surface roughness of the two systems. Once a steady state erosion condition is established, the erosion performance of the two systems, at 30° impact, is much closer. Previous studies using this erosion rig at 90° impact angle [3] has shown that the erosion rate of a plasma sprayed and EB PVD TBC differ by a factor of 7 at room temperature with the EB PVD TBC having the lower erosion rate. For glancing angle impact, the erosion rate of EB PVD TBCs is known to depend on the difference in orientation of the impacting particle on the column growth, with a significant increase in the erosion rate for open EB PVD structures when this difference in orientation exceeds 58° [4].

Table.1 Erosion rates of reference systems (top coat/ bond coat / Hastelloy X) and (bond coat/ Hastelloy X).

	Reference TBCs : YSZ top coat/Bond coat/ Hastelloy X		Reference bond coats / Hastelloy X	
Erosion rate [g.kg-1]	YSZ EBPVD /NiAl	YSZ Plasma /NiCrAlY	NiAl/Hastelloy X	NiCrAlY/Hastelloy X
Short time exposure	2.8	22.9	2.7	10.7
Long time exposure	2.3	4.6	0.6	1.2
Ra [µm]	1.5	~13	0.9	10<Ra<15

For completeness the erosion performance of the bond coat systems were also studied (see table 1 and Fig. 5). These first tests show that erosion rates are lower than for the ceramic systems. A 30°

impact angle is likely to produce the highest erosion rates for these metal systems. Again a difference is obtained between the erosion behaviour of the NiAl bond coat (by CVD) and the plasma sprayed MCrAlY system.

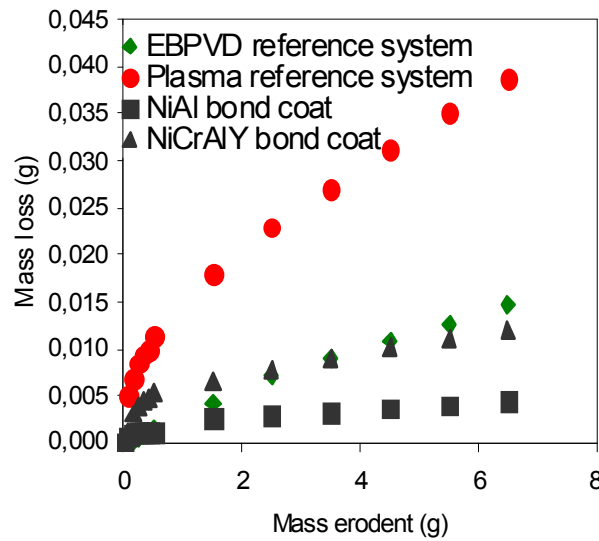


Fig. 2. Mass loss of the reference samples vs. the mass of the erodent projected at their surface.

**Characterization of sol-gel samples.** For the sol-gel TBCs the thickness of the TBC produced depends strongly on the roughness of the bond coat to be TBC coated. For the system (NiCrAlY/Hastelloy X), the thickness of the sol-gel top coat (YSZ SG) lies between 5 $\mu$ m and 25 $\mu$ m (5 $\mu$ m on high points, 25 $\mu$ m in hollows). This relates to the dip-coating method of production which can fill the hollows. This is not the case on the NiAl bond coat, which is less rough, the thickness of the YSZ SG layer is about 8 $\mu$ m.

Table 2 presents the erosion rates of the YSZ SG top coats on the NiCrAlY/ Hastelloy X and NiAl/ Hastelloy X bond coat systems. As the sol-gel samples are thinner than the traditional TBCs, they were only exposed to the erodent particles for a short time of exposure.

Table.2 Erosion rates of samples obtained using the sol-gel process.

Erosion rate [g.kg <sup>-1</sup> ]	Sol-gel top coat	
	YSZ SG / NiAl/Hastelloy X	YSZ SG / NiCrAlY/Hastelloy X
Short time exposure	10.8	13.5
Long time exposure	-	-
Ra [ $\mu$ m]	3.4	10<Ra<15

The first thing that one can see from the results of the erosion tests of the thin sol-gel TBCs compared to their respective bond coats, shown in Fig. 3 and 4, is that as for the standard TBCs their erosion rates are higher than that of the bond coats. It also appears as though the bond coat affects the erosion rate, since the sol-gel TBC on the aluminide bond coat is slightly lower than the TBC on the sprayed bond coat. This is most likely to do the differences in the roughness of the bond coats. There are a number of issues to consider when examining these results. The most important is the size of the impacting particle relative to the thickness of the coating. In the case of these sol-gel TBCs the ceramic layer is very thin in the order of 10-20% of the diameter of the impacting particles. This means that the bonding of the TBC to the bond coat will play a significant role in the

erosion resistance of the coating while the ‘bulk’ properties will have less of an effect. The other important point is that due to the aforementioned the sol-gel TBCs do not attain a steady state erosion rate.

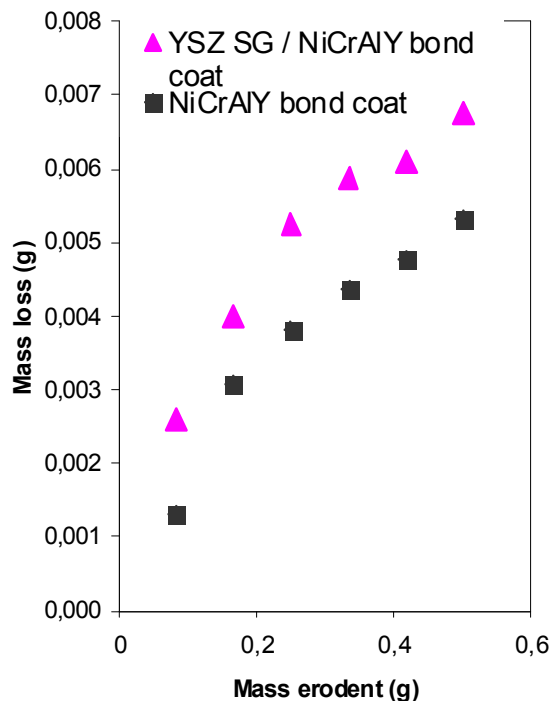


Fig. 3. Mass loss of the YSZ SG top coat deposited on the NiCrAlY bond coat vs the mass of the erodent projected at their surface.

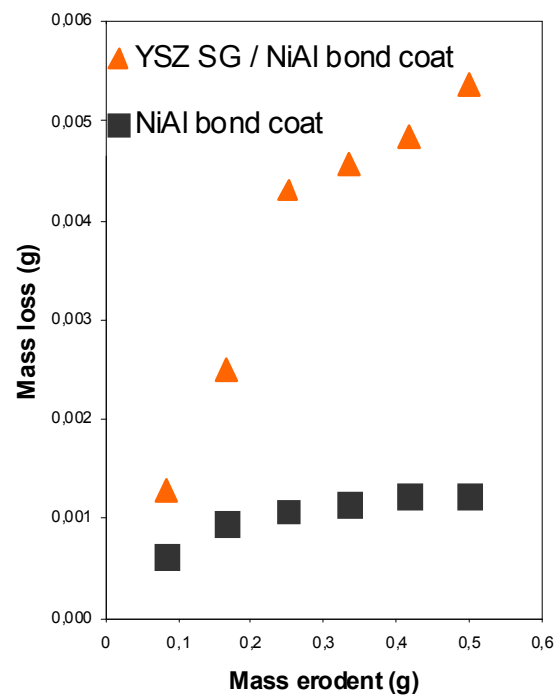


Fig. 4. Mass loss of the YSZ SG top coat deposited on the NiAl bond coat vs the mass of the erodent projected at their surface.

**Characterization of the aged samples.** The systems (YSZ SG/ bond coat/ Hastelloy X) were aged using an isothermal treatment at 1100°C, for 100 h. They were then characterized under the same conditions as it was previously described. Table 3 presents the erosion rates of the as deposited sample (noted AD) and those obtained for aged samples (HT for High Temperature). Fig. 5 shows the evolution of the mass loss of coating versus the mass of impacting erodent.

Aging was found to increase the erosion rate of both sol-gel deposited TBC systems. This is similar to previous studies on EB PVD TBCs, where the erosion rate increased by a factor of two following aging for 100 h at 1100°C for a standard EB PVD TBC system[5]. Although in the case of the sol-gel TBC a different mechanism will be operating which is most likely dependent on the formation and growth of the TGO layer.

Table.3 Erosion rates of the aged samples (HT) prepared using the sol-gel process (YSZ SG), comparison with the as deposited samples (AD).

Erosion rate [g/kg]	YSZ SG / NiAl/Hastelloy X		YSZ SG / NiCrAlY/Hastelloy X	
	AD	HT	AD	HT
Short time exposure	11	16	14	19

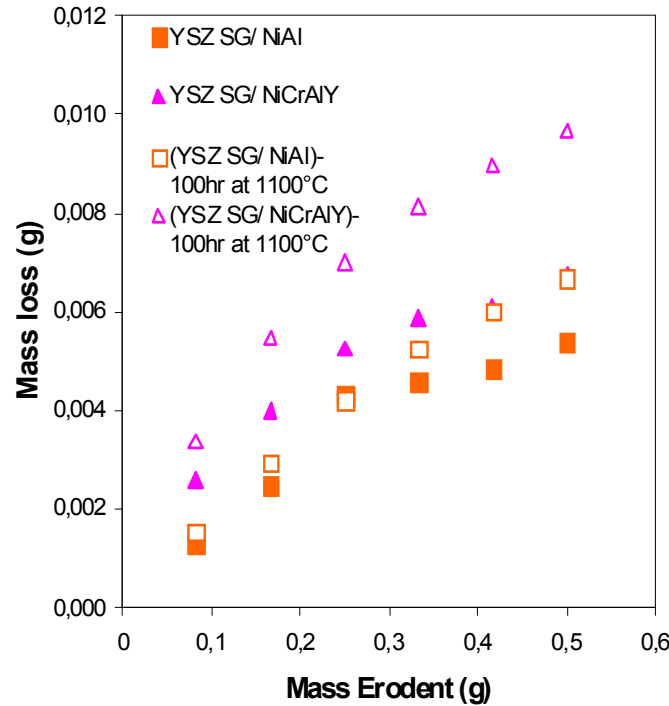


Fig. 5. Mass loss of the aged 100 h at 1100°C and not aged YSZ SG samples versus the mass of the erodent projected at their surface.

**Summary of the erosion tests.** This study has shown that, unfortunately, the thin sol-gel TBCs have extremely poor erosion resistance to impact by large alumina particles. The erosion rate was found to be 4-5 times higher than standard EB PVD TBCs and 2-3 times higher than standard PS TBCs. Furthermore the erosion rate was found to increase 30-40% with aging. Considering the large size of the impacting particles compared to the thickness of the coatings this finding is not surprising. It is most likely that the bonding between the sol-gel TBC and the bond coat has the biggest influence on the erosion rate.

### Cyclic oxidation testing

**Description of the samples.** YSZ sol-gel coatings were deposited on the ((Ni, Pt)Al/ AM1) and (NiCrAlY/ Hastelloy X) systems. On the (Ni, Pt)Al bond coat, the thickness of the YSZ layer is equal to 20μm. For the (NiCrAlY/ Hastelloy X) substrate, the thickness of the sol-gel deposit lies between 18μm and 80μm, Micrographs of both systems are illustrated in Fig. 6.

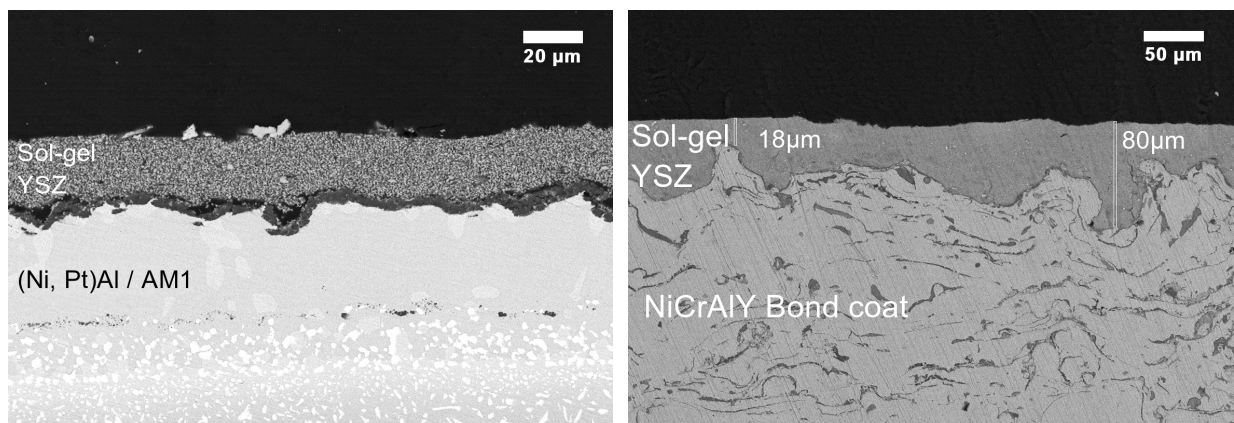


Fig. 6. Cross section (SEM) of the YSZ SG coating deposited on the ((Ni, Pt)Al/ AM1) (left) and (NiCrAlY/ Hastelloy X) (right) systems.



**Performances of the YSZ SG coatings during the cyclic oxidation test.** The delamination of the YSZ coating deposited on the ((Ni, Pt)Al/ AM1) systems is observed after 200 cycles (Fig. 7). It can be observed from the photographs in Fig. 7 that a part of the YSZ SG layer is still present on the surface of the substrate. Nevertheless, this level of degradation implies the end of the cyclic oxidation test. The delamination of the ceramic top coat was initiated at the edge of the sample and it was observed to propagate progressively across the whole YSZ layer. From a microscopic point of view, Fig. 8 presents the cross section of the sample after the cyclic oxidation test. One can note that the YSZ layer is not fissured. Moreover, the delamination of the sol-gel thermal barrier coating takes place at the interface between the TGO and the ceramic top coat, with some residual YSZ agglomerates are still bonded to the TGO (Fig. 8).

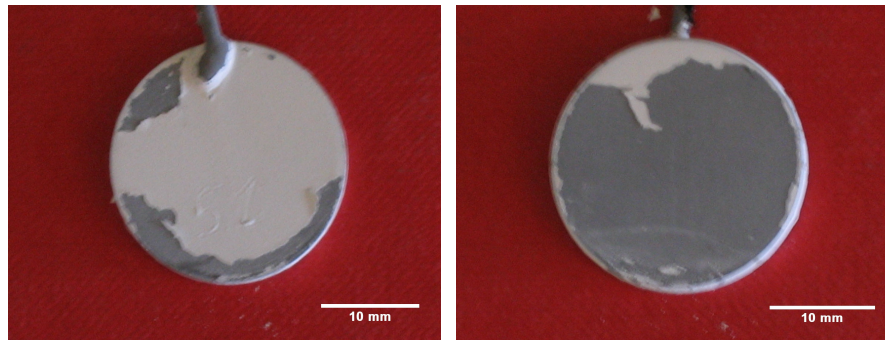


Fig. 7. Images of (YSZ SG/ (Ni, Pt)Al/ AM1) sample after 200 cycles of 1 h at 1100°C (recto : left ; verso : right).

The results obtained with the (YSZ SG/ NiCrAlY/ Hastelloy X) sample are different. Only partial delamination of the YSZ coating is observed after 740 cycles of 1 h at 1100°C (Fig. 9). This really encouraging result could be explained by the mechanical anchoring of the sol-gel YSZ deposit onto the surface undulations of the NiCrAlY bond coat (Fig 9b). As it was the case for the erosion tests, the roughness of the NiCrAlY bond coat makes possible to confine the sol-gel deposit which allows to reach better mechanical performances.

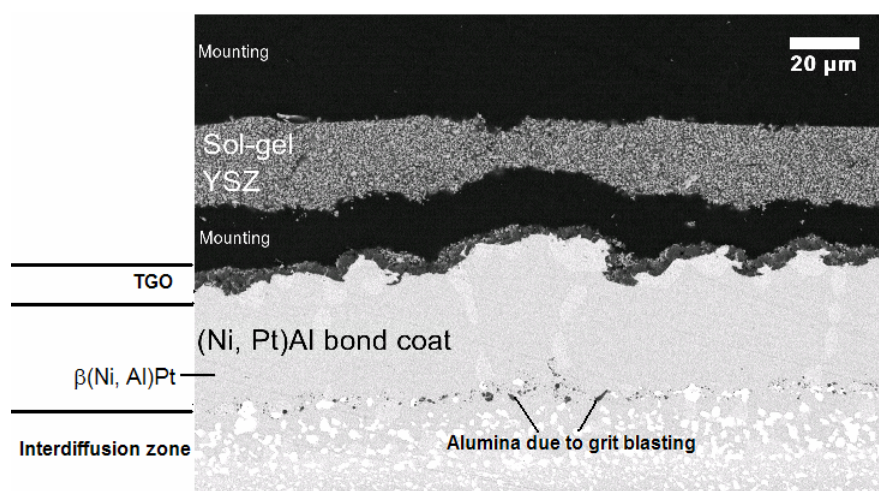


Fig. 8. Cross section (SEM) of the (YSZ SG/ (Ni, Pt)Al/ AM1) sample after 200 cycles of 1 h at 1100°C.



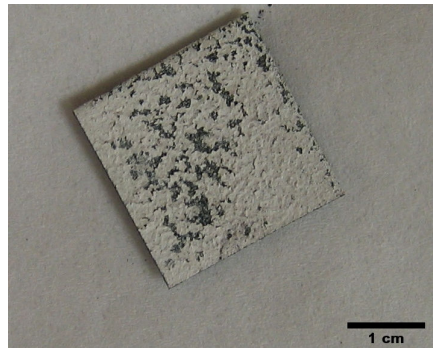


Fig. 9. (a) Photograph of the surface of the system (YSZ SG/ NiCrAlY/ Hastelloy X) after 740 cycles of 1 hour at 1100°C.(b) cross section (SEM) showing the anchoring of the YSZ SG.

### Conclusions and perspectives

The present paper presents the first erosion and cyclic oxidation tests performed on thin YSZ top coats fabricated using the sol-gel route associated with the dip-coating technique. It has been shown that the cyclic oxidation performances of the random orientated ceramic, obtained at low temperature ( $<1200^{\circ}\text{C}$ ) by this soft chemistry process, are better when the ceramic layer is deposited on a rough bond coat (NiCrAlY bond coat in this case), however the erosion resistance was found to be better for TBCs deposited onto the smoother (Pt,Ni)Al bond coat.

Recent progress in the development of the protocol allows thicker random orientated YSZ layers to be produced. It will be necessary in the future to compare the thermomechanical performances of sol-gel TBCs with the traditional ones prepared by EB-PVD and plasma sprayed processes, keeping an equivalent thickness of the ceramic top coat.

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